Auger Electron Angular Distributions from Surfaces: Forward Focusing or Silhouettes?

The recent article by Douglas G. Frank et al. (1) contains a number of conceptual errors that undermine both the data interpretation and the conclusions. The authors base their analysis on an erroneous set of notions about electron atom scattering. At the center of their misunderstanding is the statement that Auger emission events are uncorrelated and therefore cannot undergo the “formation of plane waves required for efficient diffraction.” They then go on to imply that photoemission events are correlated and that the resulting photoelectrons can thereby undergo coherent diffraction. In point of fact, both Auger emission and photoemission events are uncorrelated. Also, correlation between Auger or photoemission events is not required to realize diffraction. Diffraction is nothing more than elastic scattering and interference, and all that is required for Auger or photoelectron diffraction to be detectable in an angle-resolved measurement is that the emitter be situated in a single crystal. In such a situation, the observed angular distributions show considerable intensity modulation as a result of the interference of the unscattered wave portion and all elastically scattered wave portions at the detector point.

Frank et al. then go on to claim that the modulation is caused entirely by “shadowing” (inelastic scattering). Yet, elastic scattering cross sections show considerable angular dependence and are largely peaked in the forward direction for all but the lowest kinetic energies. In contrast, there is no convincing evidence that inelastic scattering of low to medium kinetic energy electrons at single-crystal surfaces shows any anisotropy. The primary loss mechanism is plasma excitation, which is largely delocalized. A much weaker loss mechanism is excitation of bound core states by dipole scattering, which might be expected to show some angular dependence. In making their assertions, Frank et al. tacitly ignore 10 years of successful application of elastic scattering theory to the interpretation of angle-resolved Auger and photoelectron spectroscopic data. In rationalizing their results, they also ignore the well-established fact that Auger electrons and photoelectrons of the same kinetic energy from the same specimen exhibit nearly identical angular distributions, all of which are very well predicted by elastic scattering theory (2).

So why do the data of Frank et al. show minima along interatomic directions in Pt(111)? One possibility is that their specimens were not properly oriented about the surface normal during the measurements (3). If their crystal was rotated 60° about the surface normal relative to where they thought it was, the low-energy electron diffraction pattern would not change, but the Auger intensity pattern would be inverted. If this error was made, what was interpreted to be electron intensity poking through the spaces between surface atoms would actually be forward-scattering-induced maxima along interatomic vectors. The latter interpretation is certainly much more consistent with basic principles of quantum-mechanical scattering than the proposition they forwarded.

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The physics underlying the angular distribution of Auger electrons is well understood (1), having been the subject of study for two decades. However, the recent article by Frank et al. (2) dismisses as mistaken virtually the whole of this body of work. We believe that the work of Frank et al. is mistaken, that the well-established models are the correct ones, and that the origin of this dispute is a very limited, unrepresentative data set that Frank et al. have interpreted with models that contain a mixture of gross oversimplification and error.

The crux of the interpretation by Frank et al. is that atoms cast shadows so that Auger intensities are weak along interatomic directions. As a general proposition this is false. For Auger electrons with a kinetic energy below ~100 eV, the angular distribution of the intensity is observed experimentally to be a strong function of kinetic energy. The directions in which high and low intensities are observed are different for Auger electrons with different kinetic energies (3), and this is incompatible with the model proposed by Frank et al. Moreover, the Auger electrons from different materials with a common crystal structure give different angular distributions (4). These results are due to complicated diffraction phenomena involving multiple elastic scattering of the emitted Auger electron, effects that are highly energy dependent. Therefore the approach to determining surface structure that Frank et al. suggest has no general validity.

At kinetic energies of a few hundred electron volts and above, Auger angular distributions are well known to exhibit enhanced intensities along interatomic directions because of forward scattering (or forward focusing). In the past decade this effect has been developed into a useful tool for surface structural determination (5).

Other errors in this paper are too numerous for a complete list here, but among the most egregious are the claims that only 4% of the scattering events are elastic and that it is the bound electrons (rather than the total atomic potential) that scatter an incident Auger electron.

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Frank et al. (1) claim that Auger electrons emitted from atoms in deeper layers have angular profiles that peak along the spaces between surface atoms. These authors explain their result as surface atoms casting shadows or silhouettes on the emission from deeper layer atoms. In other words, the surface atoms block the transmission of Auger electrons along interatomic directions. They claim that the shadowing effect is generally valid and that this effect presents a direct method for imaging surface atomic structure. In contrast, previously published
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